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published in
Physical Review A. Atomic, Molecular and Optical Physics
1987

DOI (link to publisher)
10.1103/PhysRevA.36.4081

document version
Publisher's PDF, also known as Version of record

Low autoionization rates in the $[5dnf]$ $J=4,5$ doubly excited series in Ba I

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(Received 16 March 1987)

In a cw laser spectroscopic experiment using a collimated atomic beam of barium we have excited the $5dnf$ $J=4$ and 5 autoionizing series. In transitions from the metastable $5d^2\,{}^1G_4$ level inherently narrow autoionization linewidths, down to a Doppler-limited value of 10 MHz, have been observed. Assignment of all observed $5dnf$ transitions has been made.

In recent years barium, with its two valence electrons simultaneously excited in multistep excitation using pulsed dye lasers, has been the subject of several investigations as to its autoionizing states.[1-3] The first experiments on barium autoionizing states with narrow-band cw dye-laser radiation have recently been performed by Neukammer *et al.*[4] They observed line narrowing and thus stabilization of some of the $5d_{3/2}nd_{3/2}$ $J=0$ states (down to 6 MHz), caused by configuration interaction with perturbing states. Van Woerkom *et al.*[5] measured the $5d_{3/2}26d_{3/2}$ $J=0$ lifetime to be 190 ns implying a 0.84-MHz autoionization-induced linewidth.

We report the observation of $5dnf$ $J=4$ and 5 series of autoionizing states in barium converging to the second $5d_{3/2}$ (at 46 908.75 cm^{-1}) and third $5d_{5/2}$ (at 47 709.73 cm^{-1}) ionization limit. Also levels of the $5dng$ and $5dnh$ configuration are observed. Most of the transitions show overall small linewidths [down to 10 MHz full width at half maximum (FWHM)], justifying the use of a stabilized cw dye laser. The states are excited from the metastable $5d^2\,{}^1G_4$ level at 24 696.278(6) cm^{-1}.[6] The $6s\epsilon l$ continua are not excited in this way. The $J=1$ $5dnf$ states have already been observed by Garton and Tomkins.[7]

Only a brief description of our experiment is given here. More details can be found elsewhere.[6,8] A collimated beam of barium is orthogonally intersected by the output beam of a single-mode ring dye laser (Spectra Physics model 380D) using the dye Stilbene 3. An electron multiplier to detect electrons escaping from the autoionizing atoms is positioned directly above the intersection point. Electrons from $5d_{5/2}nf$ states autoionizing to a $5d_{3/2}\epsilon l$ state are so slow (<0.1 eV) that most of them do not reach the electron multiplier. To observe these electrons a small extraction voltage has to be applied. In this way it is possible to discriminate between the two possible Ba II end states, $6s_{1/2}$ and $5d_{3/2}$. The $5d^2\,{}^1G_4$ level is populated by the hot tungsten heating wire in front of the barium-filled tantalum oven. The minimum width of the observed spectral lines is 10 MHz (FWHM), which is about equal to the residual Doppler effect from atomic beam divergence and focusing of the laser beam.

In transitions from the $5d^2\,{}^1G_4$ level both $5d_{3/2}nf$ and $5d_{5/2}nf$ multiplets may be excited. In the $5d_{3/2}nf$ multiplet we expect two levels with $J=3$, two with $J=4$, and one with $J=5$ which can only ionize to $6s\epsilon l$ continua. In the $5d_{5/2}nf$ multiplet we expect the excitation of two levels with $J=3$, 4, and 5 which ionize to $6s\epsilon l$ and, for $n>11$, also to $5d_{3/2}\epsilon l$ continua. First we will turn our attention to the states below the $5d_{3/2}$ ionization limit.

A typical spectrum of a $5d_{3/2}$ nf multiplet shows one $J=5$ and two $J=4$ states ($n=41$ in Fig. 1). The states

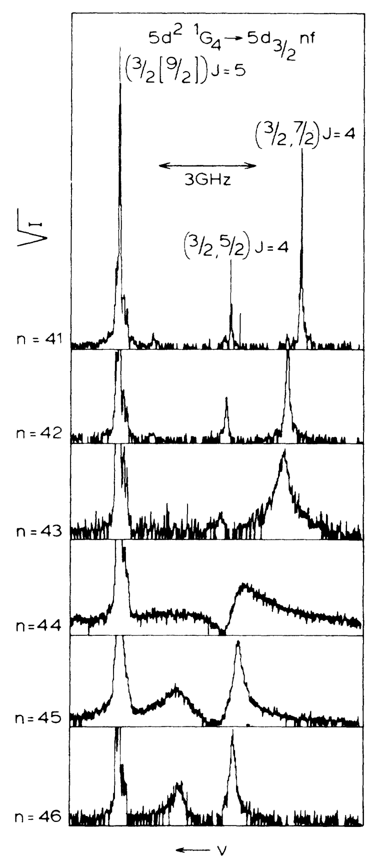

FIG. 1. The $5d_{3/2}nf$ ($n=41$–46) multiplets. The $J=5$ state has a jK coupling assignment, the $J=4$ states have jj coupling assignments.

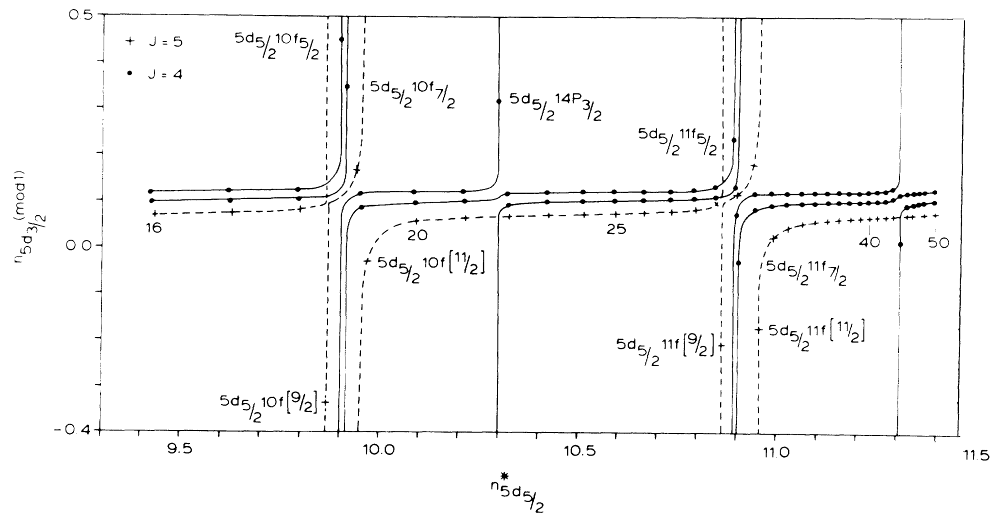

FIG. 2. Lu-Fano plot of the $5dnf$ and $5dnp$ $J=4$ and 5 states below the $5d_{3/2}$ ionization limit. The lines drawn are preliminary.

are sufficiently stable to enable a J value assignment with the help of the Stark splitting of the lines. At low electric field strengths (50V/cm at $n=20$) the $J=5$, and for some n values the $J=4$ $5d_{3/2}nf$ states, exhibit a regular quadratic Stark splitting. The number of observable $|m|$ components gives a value for J, which may be verified with the relative separations between the $|m|$ components as derived from the well-known formulas describing the quadratic Stark effect. This also results in values for the polarizability parameters α_0 and α_2. The different m components in general have different linewidths; $|m|=0$ components have the largest width. At higher n values (>25) the two $J=4$ states are so close that the lines split in a more complicated pattern due to fine-structure mixing. Since the $5dnf$ states have a small quantum defect (approximately 0.06 for the $5d_{3/2}nf_{7/2}$ $J=5$ series) and thus are nearly hydrogenic the linear Stark effect (Stark manifold) may be observed at higher field strengths. A study of the bewildering variety of electric-field-induced effects in this configuration is in progress in our laboratory. The J assignment based on the Stark effect is confirmed by the observation of series interactions and by oscillator strength considerations.

Relative strengths of the two $J=4$ and one $J=5$ lines show significant variations over the series, but for each value of n the $J=4$ line in the middle, see Fig. 1, is weakest. The transition to the $J=5$ state is always by far the strongest. Lines which could possibly be assigned transitions to $5d_{3/2}nf$ $J=3$ states have only been observed for some values of n close to perturbing $5d_{5/2}nf$ states. Transitions to $J=3$ states from $5d^2\,{}^1G_4$ may be expected to be much weaker than transitions to $J=4,5$ states. A significant difference in character between the $5d_{3/2}nf$ $J=3$ and the $J=4,5$ series is that the $J=3$ series may interact with the $5d_{3/2}np_{3/2}$ $J=3$ series. The $5dnp$ series show large linewidths; e.g. we measure the $5d_{5/2}14p_{3/2}$ state, located below the $5d_{3/2}$ ionization limit, to have a 80-GHz linewidth. This implies that even weak configuration interaction between the $5dnf$ and $5dnp$ $J=3$ series results in significant broadening of the $5dnf$ $J=3$ lines. As we excite the atoms with narrow-band laser radiation (1 MHz) this will lead to a corresponding decrease in signal strength. Combined with the relatively low oscillator strength this explains the difficulty in observing these levels. Near perturbing $5d_{5/2}nf$ $J=3$ states, oscillator strength and linewidth variations can take place[4,8] which will enable the observation of isolated $5d_{3/2}nf$ $J=3$ states.

A Lu-Fano plot of the $5dnf$ $J=4$ and 5 series below the $5d_{3/2}$ ionization limit is given in Fig. 2. It clearly

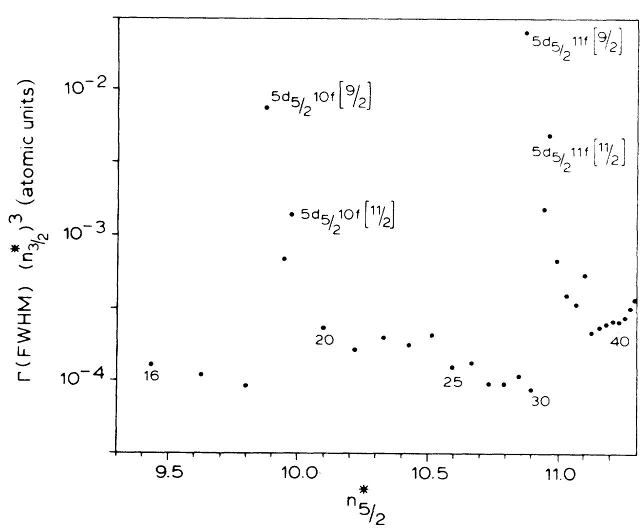

FIG. 3. The value of $(n^*)^3\Gamma$ (FWHM) of the $5d_{3/2}nf[9/2]$ $J=5$ and perturbing states.

shows the interactions between series converging to the $5d_{3/2}$ and $5d_{5/2}$ limit. Perturbing states are identified by their large linewidth due to lower l and/or n quantum numbers. These series interactions lead to considerable variations in linewidths along the $5d_{3/2}nf$ series.

The two $5d_{3/2}nf$ $J=4$ ($n=16-50$) series are perturbed by two $5d_{5/2}nf$ $J=4$ ($n=10,11$) series and one $5d_{5/2}np_{3/2}$ $J=4$ ($n=14$ and 15) series. The perturbations are well localized in the Lu-Fano plot. However, the linewidths of the two $5d_{3/2}nf$ $J=4$ transitions sometimes become as large as the separation between the two states. As a consequence, several interference effects may be observed. A nice example is the $n=41-46$ interval where the $5d_{5/2}15p_{3/2}$ $J=4$ state perturbs the $5d_{3/2}nf$ $J=4$ states as shown in Fig. 1. At $n=44$ one of the $J=4$ states is broadened by and lies on top of the $5d_{5/2}15p_{3/2}$ $J=4$ perturbing state, giving rise to a Fano profile. The other $J=4$ state is shifted to higher energy because of the interaction with the perturber (see Fig. 2).

Next we will consider the three $J=5$ series. The levels below the $5d_{3/2}$ limit of these series have linewidths which are much smaller than their separations. The linewidths of the $J=5$ states are shown in Fig. 3 as a function of $n^*_{5/2}$ (effective quantum number with respect to the $5d_{5/2}$ limit). The effect of stabilization just below and broadening above the perturbing state is observed.[4,9] The overall narrow linewidth is remarkable. The mean values of $(n^*)^3\Gamma$ half-width at half maximum (HWHM) for the $5dnf$ $J=5$ states below the $5d_{3/2}$ limit are 500 GHz for the $5d_{3/2}nf$ $J=5$ series, and 650 and 4100 GHz, respectively, for the two $5d_{5/2}nf$ $J=5$ series. These values are a factor of 10–100 lower than the mean values for the $5d_{3/2}nd$ series[4] also ionizing to the $6s$ ion state. This large difference can be understood qualitatively by noting that in $5dnl$ series $5dnf$ is the first series with a non-core-penetrating orbit.

Another remarkable fact is that the $5d_{5/2}nf$ $J=5$ series with quantum defect 0.04 perturbs the $5d_{3/2}nf$ series much more strongly than the other $5d_{5/2}nf$ $J=5$ (at quantum defect 0.13). This observation is confirmed by spectra of the $5d_{5/2}nf$ $J=5$ configuration above the $5d_{3/2}$ limit where ionization to the $5d_{3/2}$-ion state is possible. Figure 4 shows the $5d_{5/2}50f$ transition observing all electrons (upper part, ionization to both $5d_{3/2}$ and $6s$ end states) and only the 0.5-eV electrons (lower part, ionization to the $6s$ ion state). We can easily recognize the lines by their quantum defect as found below the $5d_{3/2}$ limit. The $5d_{5/2}nf$ $J=5$ state at defect 0.13 shows up as a narrow line, and mainly ionizes to the $6s$ ion state thereby confirming that the interaction between this series and the $5d_{3/2}$ channel is very small. The value of $(n^*)^3\Gamma$ (HWHM) for this series changes from 4100 GHz at $n=11$ below the $5d_{3/2}$ ionization limit to 5400 GHz above this limit (at $n=20$, 30, 35, 40, and 50). This is a small change from which it can be deduced that the interaction of this $5d_{5/2}nf$ series with the $6s\epsilon h$ $J=5$ channels is significantly larger than with the $5d_{3/2}nf$ $J=5$ channel. The $5d_{5/2}nf$ $J=5$ state at defect 0.04 shows up in Fig. 4 as a broad line and mainly ionizes to the $5d_{3/2}$ ion state. The value of $(n^*)^3\Gamma$ (HWHM) for this series changes form 650 GHz to 35 000 GHz over the $5d_{3/2}$ ionization limit showing this $5d_{5/2}nf$ $J=5$ series has the strongest interaction with the $5d_{3/2}nf$ $J=5$ series. The narrow and weak lines in Fig. 4 on the left side are identified as transitions to $5d_{5/2}50h$ and $5d_{5/2}50g$ states. The latter can be excited due to the presence of a small electric field (upper spectrum).

To further assign the observed lines J. E. Hansen of the Zeeman laboratory of the University of Amsterdam has performed *ab initio* Hartree-Fock calculations[10] including spin-orbit interactions for some $5dnf$ $J=4$ and 5 states including ionization to $6s$ and $5d$ continua. It appears that the $J=5$ states are best described in jK coupling and the $J=4$ states in jj coupling. The agreement between calculated and observed splittings and relative linewidths is sufficiently good to warrant the assignments of the states as indicated in the figures.

The narrow $5d_{3/2}nf$ lines ($n>30$) and the $5d_{5/2}nf[9/2]$ $J=5$ ($n>40$) lines are surrounded by small peaks which are identified as hyperfine structure peaks. The ^{138}Ba-^{136}Ba isotope splitting in the $5d^2\,{}^1G_4 \rightarrow 5dnf$ transition can be neglected.

Multichannel quantum-defect theory (MQDT) analyses of the $5dnf$ series are now in progress in our laboratory. These analyses together with the measured transition wavelengths and level energies will be published in the near future.

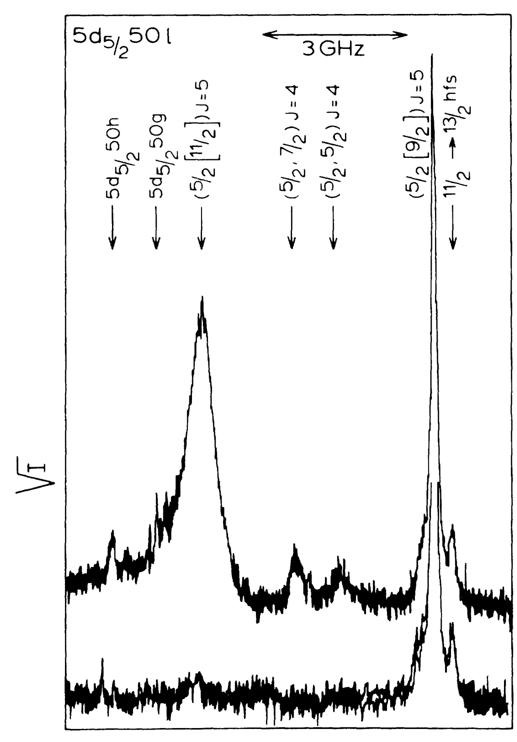

FIG. 4. The $5d_{5/2}50f$ multiplet. The lower spectrum was taken at zero electric field showing only the 0.5-eV electrons. The upper spectrum at 170-mV/cm electric field strength showing all electrons.

We thank J. E. Hansen of the Zeeman laboratory of the University of Amsterdam for performing the calculations and stimulating discussions. We also like to thank R. J. de Graaff, J. Bouma, C. T. W. Lahaye, and W. Vassen for their assistance during the experiments and gratefully acknowledge financial support from the Foundation for Fundamental Research on Matter (FOM) and the Netherlands Organization for the Advancement of Pure Research (ZWO).

[1]S. M. Jaffe, R. Kachru, H. B. Linden van den Heuvel, and T. F. Gallagher, Phys. Rev. A **32**, 1480 (1985).

[2]P. Camus, M. Dieulin, A. El Himdy, and M. Aymar, Phys. Scr. **27**, 125 (1983).

[3]R. M. Jopson, R. R. Freeman, W. E. Cooke, and J. K. Bokor, Phys. Rev. A **29**, 3154 (1984).

[4]J. Neukammer, H. Rinneberg, G. Jönsson, W. E. Cooke, H. Hieronymus, A. König, K. Vietzke, and H. Spinger-Bolk, Phys. Rev. Lett. **55**, 1979 (1985).

[5]L. D. Van Woerkom, J. G. Story, and W. E. Cooke, Phys. Rev. A **34**, 3457 (1986).

[6]W. Vassen, E. A. J. M. Bente, and W. Hogervorst, J. Phys. B **20**, 2383 (1987).

[7]W. R. S. Garton and F. S. Tomkins, Astrophys. J. **158**, 1219 (1969).

[8]B. H. Post, W. Vassen, and W. Hogervorst, J. Phys. B **19**, 511 (1986).

[9]M. Aymar, J. Phys. B **18**, L763 (1985).

[10]R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, 1981).